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Optimization of Temperatures Heating Melt and Annealing Soft Magnetic Alloys

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Abstract. Taking into account the concept of the quasi-chemical model of the liquid micro-non-uniform composition and the research made on the physical properties of the Fe-based melts being crystallized, the unique technology of the melt time-temperature treatment has been developed. Amorphous ribbons produced using this technology require optimal annealing temperatures to be specifically selected. Temperature dependences of the kinematic viscosity of a multicomponent $Fe_{72.5}Cu_1Nb_2Mo_{1.5}Si_{14}B_9$ melt have been studied. A critical temperature is detected above which the activation energy of viscous flow of the melt changes. Upon cooling the overheated melt, the temperature curves of the kinematic viscosity become linear within the given coordinates. In amorphous ribbon produced in the mode with overheating the melt above the critical temperature, the enthalpy of crystallization grows, the following heat treatment results in an increase in magnetic permeability.

1. Introduction

The concept of the quasi-chemical model of the liquid micro-non-uniform composition is being developed in our Research Center [1]. According to it, the metal melt consists of space areas (groups, cybotaxis or clusters) within which the atom arrangement is characterized by certain ordering – short-range order. Due to moderately intense particles' heat movement, the clusters have no clear-cut boundaries: the atoms' ordered arrangement is being continuously replaced by another one, moving away from the cluster's core. For the same reason, the time of the given cluster's existence is limited and depends upon the chemical bonds' energy. At the same temperature, it is possible for two or more cluster ordering types to co-exist.

Soft magnetic nanocrystalline materials are distinguished by low coercivity and high magnetic permeability magnitudes, which are provided by nanocrystalline, structure [2]. The fabrication of these materials includes three stages. At the first stage, an alloy of a given composition is melted. Then, using ultrarapid quenching, amorphous metallic ribbon about 25 μm in thickness is produced. At the third stage, in the course of heat treatment, in the ribbon there forms an optimum nanocrystalline structure.

The effect of the temperature of preparation of the melt prior to casting, which is aimed at creation in the iron-based alloys of amorphous structure, was investigated in a number of works [3–6]. It was shown that, with increasing temperature of the melt prior to casting, the ribbon becomes more plastic, its electrical resistance and crystallization temperature grow, and its density lowers. On the other hand, the increased temperature of the melt deteriorates the quality of the ribbon surface and efficiency of its production. Therefore, it is important to find an optimum mode of preparation of the melt prior to casting. As a characteristic point for a multicomponent melt, a critical temperature T_k can be specified

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[6], above which the properties of the melt irreversibly change upon cooling. Irreversibility manifests itself in a hysteresis of kinematic viscosity, surface tension, and other properties of the melt. The hysteresis can arise as a result of breaking of interatomic bonds which control the short-range order in the melt.

In the paper, the results of measurements of the viscosity of the $Fe_{72.5}Cu_1Nb_2Mo_{1.5}Si_{14}B_9$ melt are presented. The parameters of process of crystallization and magnetic properties of ribbons produced upon different modes of preparation of the melt prior to casting are investigated.

2. Experimental

The main method used in this work – a torsional oscillatory method of measurement of kinematic viscosity of metal melt. The main measured size – logarithmic decrement of attenuation of torsional fluctuations of a crucible with metal melt. In a consequence logarithmic decrement of attenuation is recalculated to kinematic viscosity on Shvidkovsky's equations [7].

Distinctive features of the used experimental plant:

Measurement of logarithmic decrement is completely automatic [8]. It excludes influence of "a human factor" on results of measurements. Besides, the automatic mode allows to receive the maximum quantity of data for a unit of time. One measurement of logarithmic decrement of attenuation of fluctuations takes about 40 seconds.

The original system of an initial swing of suspended system is used. Use of this system allows to carry out a fast swing and to avoid emergence of cross fluctuations of suspended system [9].

The special software is used. It allows to use the steps counter of the computer as the counter for measurements of time. It provides high precision of measurements.

Experiments were made in the atmosphere of helium. The internal volume of experimental installation was pumped out to vacuum of $3 \cdot 10^{-3}$ mm Hg before filling of the camera with helium. The mass of a sample is about 25 grams. Diameter of a sample is about 13 millimeters. Height of a sample is about 20 millimeters. Crucibles from beryllium oxide are used. The period of fluctuations of suspended system is about 4 seconds. Heating is carried out by means of the heater made of molybdenum. Regulation of heating was carried out by means of the simistor block by a pulse-phase method. Temperature was controlled with the help of tungsten-rhenium thermocouple. Regulation of temperature was carried out automatically with use of proportional integrated differential algorithm.

$Fe_{72,5}Cu_1Nb_2Mo_{1,5}Si_{14}B_9$ alloys were melt in a vacuum induction furnace. A 25 μm thick and 10 mm wide ribbon with the amorphous structure was produced by the planar flow casting process. The ribbon was wound up onto ring-type cores with the 32 mm outer diameter and 20 mm inner diameter. The cores were subjected to annealing at T_a = 300 – 580 ℃ in the air for t = 1 hour, following the crystallization peak.

3. Results and Discussion

In the general form, the time-temperature treatment cycle is graphically represented in Fig. 1 in temperature-time coordinates. The most significant part is the establishment of the technological parameters, vis., the maximum temperature of heating T_{treat} exposure time of the melt at this temperature (t_{treat}), the temperature of the metal before exiting from the furnace t_{exit}. One variant of the procedure of determining t_{treat} is to measure the physical properties of the melt during its holding at T_{treat}, which is somewhat higher than T_k. The relaxation time of the structure and stabilization of the properties can be recommended as the value of t_{treat}. Stirring the metal proves to be effective. It enables one to decrease the holding time at the critical temperature.

The development of specific TTT technologies for a more efficient application of their potentials stipulates the changes possible in the order of alloying components and deoxidizers added, the exposure temperature of the metal, correction of the heat-treatment conditions of the parts, etc.

The typical temperature dependences of kinematic viscosity ν, electrical-resistance ρ, magnetic susceptibility χ, surface tension σ, and density d of the liquid amorphous alloys are shown in Fig. 2. It is found that the critical temperatures termed T_k are determined on basis of such data. Heating above T_k ensures a more homogenous melt state and, as a rule, causes the formation of the physical properties

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hysteresis. Overcooling ΔT is measured for this equilibrium melt. The presence of hysteresis is indicative for the differences between the initial and final melt states at T_k in the course of melting. Sometimes, the melt may be heated only up to the anomaly temperature T_{an}. The melt is held at the maximum heating temperature for a particular time during the experiment, investigating the time dependence of the properties changes at this temperature.

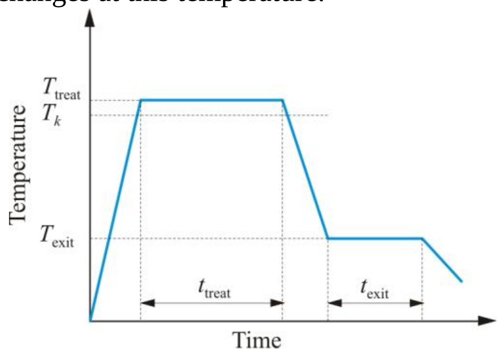

Figure 1. Thermogram of the melting of a commercial metal depicting the temperature-time treatment of the melt.

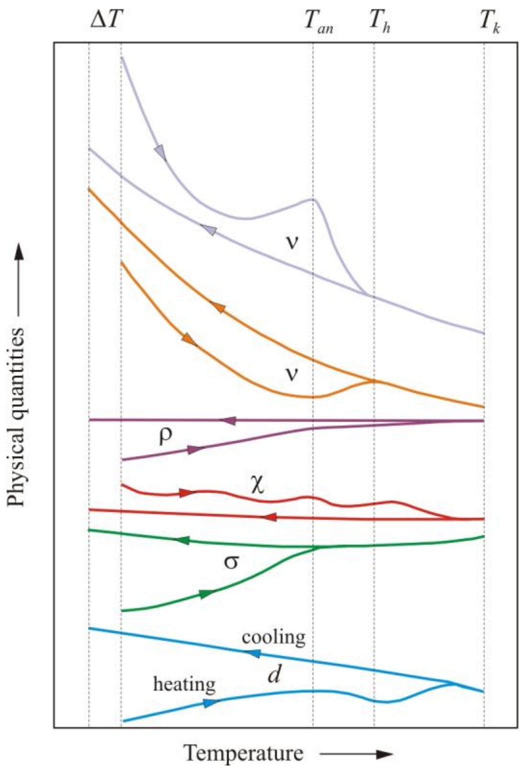

Figure 2. The typical polytherms of physical properties of amorphous alloys: ΔT is overcooling; T_{an} is the temperature of anomalies; T_h is the temperature of hysteresis; T_k is the critical temperature.

In Fig. 3, the experimental results of the viscosity measurements are presented in the form of dependence of the inverse kinematic viscosity of the melt, 1/ν, on the inverse absolute temperature, $1/T$. In the case of heating (Fig. 3a), the curve is divided into two linear portions. The change in the slope takes place at a temperature of 1490 ℃. This value coincides with the critical temperature T_k obtained earlier using the Arrhenius law. In the course of cooling (Fig. 3b), the dependence of the inverse kinematic viscosity 1/ν on the melt inverse absolute temperature $1/T$ behaves linearly with a constant slope over the whole temperature range.

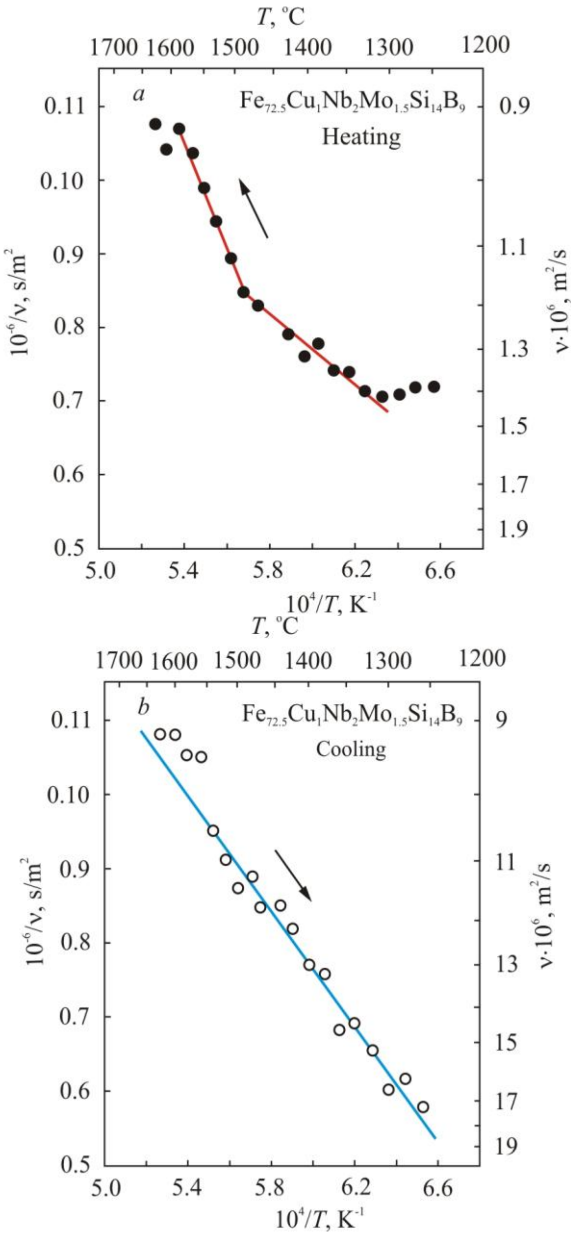

Figure 3. Dependences of the inverse kinematic viscosity of the melt 1/ν on the inverse absolute temperature 1/T in the course of heating (a) and cooling (b).

The results obtained indicate that for the multicomponent $Fe_{72.5}Cu_1Nb_2Mo_{1.5}Si_{14}B_9$ melt there is the critical temperature T_k = 1490 ℃ above which the melt can be converted into another structural state with a different magnitude of the activation energy of the viscous flow E_a. The cooling of the melt from the new structural state proceeds irreversibly. To compare two states of the melt, above and below the critical temperature, a ribbon 25 μm thick was cast from the melt prepared by two modes prior the following ultrarapid cooling. In the first mode, the melt of $Fe_{72.5}Cu_1Nb_2Mo_{1.5}Si_{14}B_9$ was heated to 1480 ℃ prior to casting a ribbon. In the second mode, the melt of the same ingot was heated to 1520 ℃, then cooled to 1480 ℃, and cast. At each temperature specified the exposure was 5 min.

Figure 4 shows the in-time changes in the temperature and initial permeability in the course of heat treatment at 550 ℃ for the cores fabricated from the ribbons that were subjected to two modes of preparation of the $Fe_{72.5}Cu_1Nb_2Mo_{1.5}Si_{14}B_9$ melt.

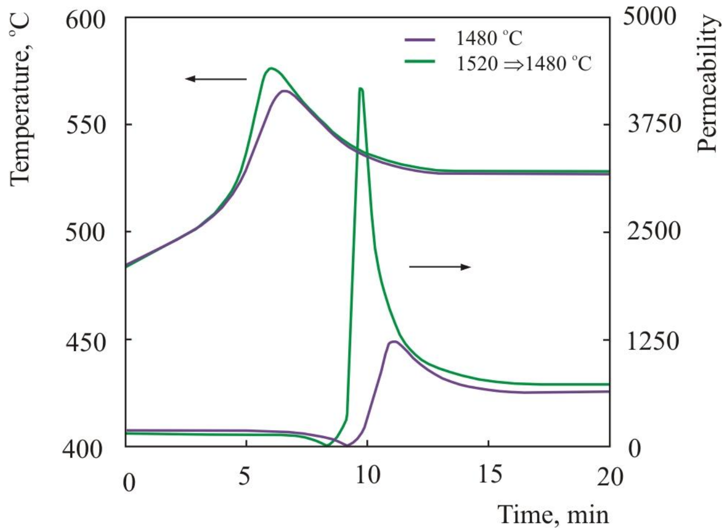

Figure 4. Temperature and permeability changes in the course of heat treatment of cores made from the ribbons undergone preparation of the $Fe_{72.5}Cu_1Nb_2Mo_{1.5}Si_{14}B_9$ melt by two modes.

The cores fabricated from the ribbons obtained by two modes of the melt preparation were subjected together to the heat treatment at the temperature 550 °C for 1 h after the crystallization peak. In Table 1, data are given on the permeability $\mu_{0.08}$ at the magnetic field strength of 0.08 A/m, maximal permeability μ_{max}, coercivity H_c of the magnetic hysteresis loop at the maximal magnetic field strength of 800 A/m. It is seen that the mode with overheating of the melt above the critical temperature provides higher magnetic properties.

Table 1 Magnetic properties of cores made from the $Fe_{72.5}Cu_1Nb_2Mo_{1.5}Si_{14}B_9$ alloy

Regime of melt preparation	$\mu_{0.08}$	μ_{max}	H_c, A/m
1480 °C	125000	882000	0.45
1520 → 1480 °C	164000	1024000	0.40

Summary

In the work, the temperature dependences of the kinematic viscosity of the multicomponent $Fe_{72.5}Cu_1Nb_2Mo_{1.5}Si_{14}B_9$ melt have been studied. A critical temperature is established above which the energy of activation of the viscous flow of the melt changes. In the course of cooling of the overheated melt, the temperature curves of the kinematic viscosity become linear within the given coordinates. In the amorphous ribbon obtained in the mode of preparation of the melt with overheating above the critical temperature, an enhanced value of the enthalpy of crystallization is found. After the heat treatments of the cores, the mode with overheating the melt above the critical temperature results in higher magnetic properties.

Acknowledgements

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